Obtaining the Volatile Oils from Wormwood and Tarragon Plants by a New Microwave Hydrodistillation Method

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In the present paper we describe an original installation for microwave hydrodistillation at low pressure that we constructed in order to obtain volatile oils from different plants. The results that we have obtained are encouraging meaning that the main components of these oils were found in correspondence with the data presented in literature. A novelty of this process is the fact that the hydrodistillation time was shorter than in the case of a classic process of hydrodistillation at atmospheric pressure. The aim of our research was to find a new and modern way of obtaining volatile oils from plants.

Keywords: volatile oils, hydrodistillation, extraction, microwave, low pressure

Volatile oils are an integral part of the class of active principles, which are complex mixtures of aliphatic, aromatic, hydrocarbons of the aldehydes, alcohols, acids, esters and, other constituents type. The composition varies according to the nature of the plant and is dependent on the development of the plant, on the climate and soil. One can often notice the difference in the composition of the volatile oil obtained from the different parts of the same plant. Most volatile oils belong to the class of the terpenoids.

Aromatic plants have been known for a very long time and the use of them in the food and perfume industry have a long history [1,2]. Many variants and systems are used to obtain volatile oils and these are based on their property of being entrained by water vapors as a result of the high vapor pressure that characterizes them. The comminution and distillation time are two important factors, which affect the essential oil yield and/or composition of aromatic plants significantly [3-5]. The water vapor entrainment has been the most commonly used process for the production of volatile oils. Microwave-assisted extraction will probably be the main technique in the near future for industrial essential oil production [6]. Nowadays the main methods of obtaining volatile oils are the classics hidrodistillation and extraction in differents solvents or the extraction with supercritical fluids [7,8].

The volatile oils making a complex mixture of substances will dissolve differently. Thus, the efficiency of the process depends on the duration of the extraction, the properties of the solvent and of the extracted substances [9,10]. Our purpose of obtaining volatile oils from plants at low pressure has been reached using an original installation.

Experimental part

Matherial and method

The procedure consists of the following operations: the raw material is brought together with the water into the distillation container, the bottom of the container is heated directly on the heat source and the mixture of oil and water vapors is collected, by condensation, into a cooling system. The distillate then passes into a collecting container.

The process of water vapor entrainment is preceded by a process of diffusion of the oil from the vegetal cells that lasts more or less depending on the location of the volatile oil in the plant and on its nature.

Apparatus

Based on the research carried out in our own chemistry laboratory and on the data presented in the specialized literature [4,6,10,11] for the production of volatile oils, we designed and built our own installation for microwave-assisted hydrodistillation at low pressure (100-200 mm Hg). A microwave generator with a modified multimode microwave device with adjustable power within the range 700-1200W was selected. A distillation flask, a refrigeration system outside the microwave generator and an adjustable vacuum pump were introduced in this [12]. The scheme of the installation is shown in the figure 1.

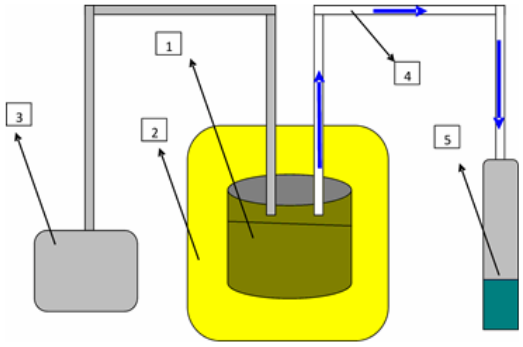

Fig. 1. Schematic diagram of the microwave-assisted hydrodistillation installation at low pressure
1. extraction balloon with vegetal material; 2. microwave generator; 3. pressure regulating device; 4. Refrigerant; 5. volatile oil collection container

Extraction of the volatile oils

The extraction of the volatile oils was carried out as follows:

- The used method of extraction: low-pressure microwave-assisted hydrodistillation process - a new installation that we conceived and designed.
- The presentation of the vegetable material: freshly harvested and subjected to a gradual drying process [5,6,9].

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All the authors had equal contribution at this original article.

Vegetable material	Reagent	Color
Wormwood	Sudan III R Solution Steimetz R Reagent	Dark red
Tarragon	Sudan III R Solution Steimetz R Reagent	Brown red

Table 1
COLOR REACTIONS FOR THE PLANTS UNDER STUDY

- The vegetation period, the geographical area: in the research that we carried out we used vegetal material from the field of experiments of the Laboratorul de Cercetare Plante Medicinale si Aromatice, Brasov (The Laboratory for the Research of Medicinal and Aromatic Plants, Brasov). The quality and composition of the plant material depend on the climatic conditions, the physico-chemical and biological properties of water and soil (influenced by excessive pollution with chemicals, pharmaceuticals, household waste, zootechnics, etc.) in the plant vegetation area [13-28].

- The harvested plant material was of from wormwood and tarragon species.

- The parameters for microwave-assisted hydro-distillation at low pressure were: p=(100-200) mm Hg; T=(53- 68) ^{0}C; contact time=24 min; dry vegetal material, ground, at φ=0.15-0.20 mm; variable power between 800-1200W.

A microscopic histochemical examination was also carried out on transversal sections of the vegetal product subjected to the analysis for the purpose of identifying volatile oils in the vegetable tissue. These identifications are made on the basis of specific color reactions of the volatile oils. Table 1 presents some data of such analyses.

In the following tables we present the analytical data obtained for volatile oils by means of low-pressure hydrodistillation. The coupling technique Gas-chromatographer-Mass Spectrometer (GC/MS) was applied under the following conditions: Type: SATURN 2100 T – ion trap detector; Carrier gas: Helium, flow rate 1 mL/min, constant pressure; Injection mode: Splitless, 2 min; Temperature of the injector: 250^{o}C; Column type: VF 5 MS, arylene modified / polydimethylsiloxane; Temperature of the program: 40^{o}C (2 min), 10^{o}C / min, 300^{o}C (10 min).

In the case of the gas-chromatographic analysis, the identification of the drops was based on the retention times of the standards for the above-described working conditions, and the determination of the quantitative composition [29] was made based on the area of the drops.

Results and discussions

We carried out 40 experiments on plant samples, twenty per each vegetal sample, having the following conditions: pressure between 100-200 mm Hg and power between 800-1200 W.

Results in Wormwood case

In figure 2 is presented the woormwood extract chromatogram and in table 2 - identification and dosage in the case of wormwood extract.

As it can be seen in our from our experimental data, we have achieved in a higher amount the following components: Sabinol=3 %, Himachalene=8.99 %, Aromadendrene=9.40 %, cis -Copaen-8-ol=14.15 % and in a smaller amount β-terpinen=0.30 %, β-pinene=0.77 %. It was also obtained a large amount of unidentified components (22.41%) that could be chemical substances as a secondary process results, being nonvolatile oils. The remaining volatile components obtained are consistent with the data in the literature.

Results in Tarragon case

In figure 3 is presented the tarragon extract chromatogram and in table 3 - identification and dosage in the case of tarragon extract.

As in the previous case, we have achieved in a higher amount the following components: Methyl eugenol=30.28 %, Asarone=24.36 %, cis-Asarone=10.46 %, representing 65 % of the entire quantity of volatile components, and in a smaller amount α-Pinene=0.30 %, β-Linalol=0.70 %. The remaining volatile components obtained are consistent with the data in the literature.

The analytical results of the volatile oils that we obtained are in accordance with the data in the literature and small differences emerged in terms of extraction efficiencies.

From the analyzed experimental data we noticed that the maximum oil efficiency was obtained when the power of the generator was adjusted to the range of 800-900 W and the pressure was adjusted to the range of 100-120 mm Hg.

Going into the Mathcad environment we used the spline functions in order to interpolate and extrapolate the experimental data. The obtained efficiency results were 847 W and 112 mm Hg.

Microwave-assisted hydrodistillation is a process that takes much less time in comparison with the classic process, maximum 30 min versus 180-210 min. It is a

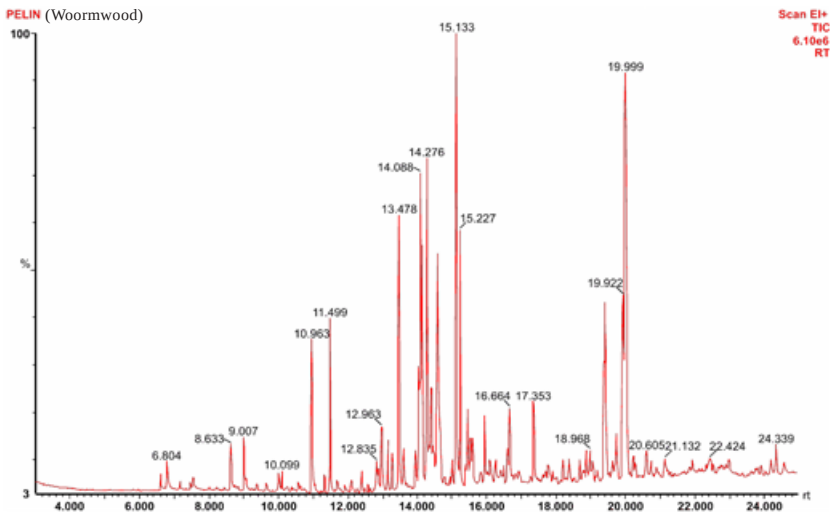

Fig. 2. Woormwood extract chromatogram

Crt. No.	Compound name	Retention time (minutes)	Mass %
1	β -terpinen	6.63	0.30
2	β-pinene	6.8	0.77
3	Camphene (2,2-Dimethyl-[3-methylenebicyclo 2.1] heptane)	7.55	0.69
4	β-Linalol	8.63	1.19
5	Thujone	9.02	1.25
6	3,6-Dimethyl-2,3,3a,4,5,7a-hexahydrobenzofuran	10.08	0.86
7	Carvone	10.96	2.44
8	Sabinol	11.5	3.00
9	Elemen	12.96	2.07
10	Linalyl isobutirate	13.14	0.92
11	Alloaromadendrene	13.48	4.52
12	Himachalene	14.10	8.99
13	Aromadendrene	14.28	9.40
14	Unidentified compound	14.59	6.60
15	Unidentified compound	15.15	10.49
16	Unidentified compound	15.51	2.97
17	Unidentified compound	15.95	2.35
18	Guaia-1(10),11-diene	16.26	0.56
19	Bisabolol	16.66	2.54
20	Chamazulene	17.37	1.79
21	Cembrene	19.73	1.11
22	cis -Copaen-8-ol	19.98	14.15
23	Other compounds	-	21.04

Table 2
IDENTIFICATION AND DOSAGE: WORMWOOD EXTRACT

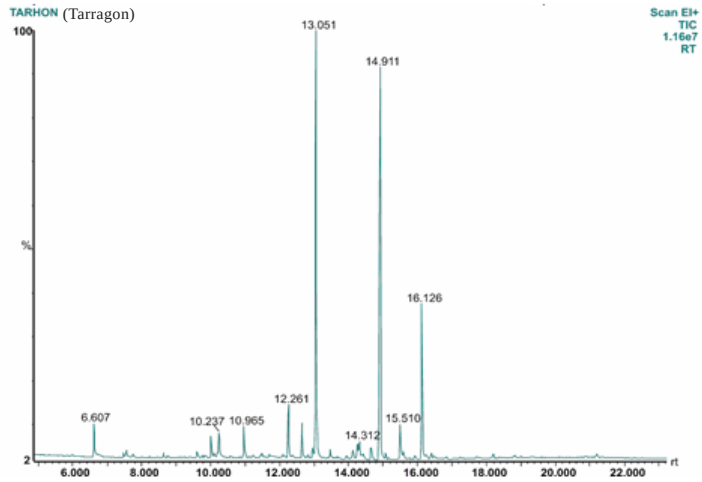

Fig. 3. Tarragon extract chromatogram

faster process and the secondary oxidation processes are diminished.

The hydrodistillation process carried out at low pressures of 100-200 mm Hg led to boiling temperatures of water of around 55ºC, which determined the volatilization of the active principles at lower boiling temperatures normal at 760 mm Hg and implicitly the hydrolysis and oxidation processes were of lower intensity.

Conclusions

The microwave processing of the plant material evidenced a number of interesting details.

The quality of the oils obtained this way is superior due to the presence of a higher percentage of principal components of the oils and to a lower percentage of artifacts, products that undergo hydrolysis and oxidation processes.

The quantity of the main components of obtained volatile oils is 5 percent bigger than in classic methods (hidrodistillation proces with Neo-Clavenger device).

Table 3
IDENTIFICATION AND DOSAGE: TARRAGON EXTRACT

Crt no.	Compound name	Retention time (minutes)	Mass %
1	Terpinen	6.62	2.81
2	Limonene	7.53	1.11
3	α-pinene	7.74	0.30
4	β-Linalol	8.63	0.70
5	p– Menthone	9.61	0.51
6	Terpine-4-ol	10.01	1.85
7	p-propenyl-anisol	10.24	2.67
8	Carvone	10.97	2.53
9	Isobornyl formate	11.48	0.55
10	Pseudoionone	11.74	0.65
11	Carane	12.26	4.31
12	Neryl acetate	12.65	2.46
13	Methyl eugenol	13.05	30.28
14	Carryophyllene	13.47	0.94
15	Cadinene	14.67	0.85
16	Asarone	14.91	24.36
17	Spathulenol	15.51	3.70
18	cis-Asarone	16.13	10.46
19	Other compounds		8.96

The maximum efficiency of the microwave-assisted hidrodistillation is obtained when parameters of power and pressure are immediately below 900 W and 140 mm Hg.

Economically speaking, the entire process takes much less time, around 30-40 min, this solution proving to be suitable for serial production.

The hydrolysis and oxidation secondary chemical processes are diminished by the given experimental conditions and also by the short duration of the hydrodistillation process carried out.

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